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## **JAPANESE / ENGLISH TRANSLATION OF**

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Soft Polyester Resin Composition and Wallpaper Made From This Composition

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#### (54) [Title of the Invention]

# Soft Polyester Resin Composition and Made Wallpaper From This Composition

#### **(57)** [Summary]

[Object] To provide a soft polyester composition that possesses improved soft polyester foamability and is less prone to the problems that develop when wallpaper or another sheet product is manufactured by means of calendering, particularly the problems encountered when such calendering is performed using a soft polyester composition with which a large amount of calcium carbonate has been admixed; and to provide wallpaper obtained using this composition.

[Means of Achievement] A soft polyester composition, characterized in that a foaming agent and 0.5 to 20 parts by weight of a (meth)acrylic acid polymer are added per 100 parts by weight of an amorphous random copolyester wherein ethylene glycol accounts for 50% or more of the diol units that constitute the copolyester.

#### [Claims]

[Claim 1] A soft polyester composition, characterized in that a foaming agent and 0.1 to 20 parts by weight of a (meth)acrylic acid polymer are added per 100 parts by weight of an amorphous random copolyester wherein ethylene glycol accounts for 50% or more of the diol units that constitute the copolyester.

[Claim 2] A soft polyester composition, characterized in that 0.5 to 20 parts by weight of a (meth)acrylic acid copolymer, 40 to 80 parts by weight of calcium carbonate, 5 to 40 parts by weight of a plasticizer comprising structural formula A, 0.01 to 5.0 parts by weight of a phosphoric acid ester lubricant represented by at least formula (I), and a foaming agent are added per 100 parts by weight of an amorphous copolyester wherein ethylene glycol accounts for 50% or more of the diol units that constitute the copolyester.

$$(-CH(R_1)-CH(R_2)-O-)_m$$
 Structural formula A (In the formula,  $R_1$  and  $R_2$  are  $C_nH_{2n+1}$ ;  $n=0$  or 1 or greater; and m is an integer of 1 or greater. When m is 2 or greater, structural formula A can also comprise alkylene oxide units with

different  $R_1$  and/or  $R_2$ , or blocks comprising different alkylene oxide units. In addition,  $R_1$  and  $R_2$  in the alkylene oxide units can be the same or different.)

$$(R-O)_n PO(OH)_{3-n}$$
 Formula (I)

(In the formula, n is an integer of 1 or 2, and R is an alkyl group, alkylphenyl group, alkylphenylpolyoxyethylene group, or alkylphenylpolyoxyethylene group.)

[Claim 3] The soft polyester composition according to claim 1 or 2, characterized in that at least 50 to 95 wt% of the polymerizable component in the (meth)acrylic acid copolymer is methyl (meth)acrylate.

[Claim 4] The soft polyester composition according to claim 1, 2 or 3, characterized in that the phosphoric acid ester lubricant is a mixture of phosphoric acid monoester and phosphoric acid diester at a ratio of 10:1 to 1:10.

[Claim 5] The polyester composition according to claim 1, 2, 3, or 4, characterized in that aluminum hydroxide or antimony trioxide is further admixed as a flame retardant.

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[Claim 6] Wallpaper manufactured using the soft polyester resin composition according to any of claims 1 through 4.

# [Detailed Description of the Invention]

[Technological Field of the Invention] The present invention relates to a polyester resin composition whose foamability has been improved as a result of admixing a (meth)acrylic acid polymer into a soft polyester resin to which foaming agent has been added; to a soft polyester resin composition with which calcium carbonate has been further admixed, particularly to a soft polyester composition with improved surface roughening, selvedge drying, foamability, drawing properties, and the like; and to wallpaper obtained using this composition.

#### [0002]

[Prior Art] Polyvinyl chloride compositions and polyvinylidene chloride compositions have good calenderability and other properties, and are therefore used to manufacture wallpaper and other sheet products. However, development of a resin composition material capable of replacing these polyvinyl chloride and polyvinylidene chloride compositions has been actively pursued in recent years. Polyester resins such as polyolefin resins, as well as many other types of resins, have been studied as this type of material. Of these, foamable wall coverings obtained using biodegradable aliphatic polyester resins (whose post-use degradability and other properties are taken into account) have been proposed during the course of development of polyester-based wallpaper and other sheet products. Numerous other techniques have also been proposed whereby polyester resins are used for foaming sheets whose thickness ranges from 0.1 to 10 mm. The use of copolyester as the polyester resin in such techniques has also been proposed, and it is also reported that a sheet that contains uniform fine air bubbles, has a high foaming ratio, and possesses thermal stability can be obtained using this type of resin (refer to JP (Kokai) 7-165968, JP (Kokai) 6-73223). However, the polyester foams cited in these two patents are intended for use in containers for instant noodles and the like, and even though it is disclosed that uniform fine cells are formed, the surfaces remain nonuniform and these foams are unsuitable for actual use from the standpoint of the characteristics required of wallpaper.

[0003] The inventors have also proposed polyolefin resins and other polyester resins as substitute resin materials. Nevertheless, even though the polyester resins proposed thus far have been used, there have been problems in that a product with sufficient properties rivaling those of decorative sheets obtained using conventional polyvinyl chloride resin and the like have not been obtained, and the above-mentioned polyester resins have not yet been suitable in terms of calenderability for manufacturing sheet products when compared to the above-mentioned polyvinyl chloride resin. Even the soft polyester composition for manufacturing wallpaper and the soft polyester composition to which large amounts of calcium carbonate were admixed in order to improve flame resistance and the like that have been proposed by the authors fail to achieve adequate foamability and pose problems in that the sheet surface becomes rough, the selvedges dry up, and elongation during calendering is poor when sheets are produced by means of calendering.

#### [0004]

[Problems to Be Solved by the Invention] Consequently, an object of the present invention is essentially to improve foamability of soft polyester resin to which foaming agent has been added, and to further provide a soft polyester composition devoid of the disadvantages that occur when wallpaper and other sheet products are manufactured by means of the calendering of the above-mentioned soft polyester sheet, particularly the problems encountered during the above-mentioned calendering of soft polyester compositions with which large amounts of calcium carbonate have been admixed; and also to provide wallpaper that is manufactured using this soft polyester composition. The above-mentioned object was attained as a result of intense studies intended to address the aforementioned problems and concerned with components that are added in order to overcome the above-mentioned disadvantages, whereby it was discovered that when the components admixed in order to improve the vacuum moldability of styrene resin compositions for foam molding are used with the above-mentioned soft polyester resin, foamability is improved and the problems encountered during calendering are overcome.

#### [0005]

[Means Used to Solve the Above-Mentioned Problems] The present invention is essentially a soft polyester composition characterized in that a foaming agent and 0.1 to 20 parts by weight of a (meth)acrylic acid polymer are added per 100 parts by weight of an amorphous random copolyester wherein ethylene glycol accounts for 50% or more of the diol units that constitute

the copolyester. The present invention also provides a soft polyester composition characterized in that 0.5 to 20 parts by weight of a (meth)acrylic acid copolymer, 40 to 80 parts by weight of calcium carbonate, 5 to 40 parts by weight of a plasticizer comprising structural formula A, 0.01 to 5.0 parts by weight of a phosphoric acid ester lubricant represented by at least formula (I), and a foaming agent are added per 100 parts by weight of an amorphous copolyester wherein ethylene glycol accounts for 50% or more of the diol units that constitute the copolyester.

$$(R-O)_n PO(OH)_{3-n}$$
 Formula (I)

(In the formula, n is an integer of 1 or 2, and R is an alkyl group, alkylphenyl group, alkylpolyoxyethylene group, or alkylphenylpolyoxyethylene group.) The present invention is preferably a soft polyester composition characterized in that at least 50 to 95 wt% of the polymerizable component in the (meth)acrylic acid copolymer is methyl (meth)acrylate, particularly a soft polyester composition characterized in that the phosphoric acid ester lubricant is a mixture of phosphoric acid monoester and phosphoric acid diester at a ratio of 10:1 to 1:10, ideally a soft polyester composition characterized in that aluminum hydroxide or antimony trioxide is further admixed as a flame retardant.

[0006] The second invention is wallpaper that has been manufactured using any of the abovementioned soft polyester resin compositions.

#### [0007]

[Embodiments of the Invention] The present invention will be described in further detail.

A. It is preferred that a copolyester resin of low crystallinity, particularly an essentially amorphous one, be used as the copolyester resin in the present invention. It is particularly preferred that the copolyester is a random copolymer. Amorphous random copolyester resins are known to have extremely stable thermal properties without forming a crystal phase during heating in the manufacturing process or during the reheating of the product. 1. Random copolyesters will first be described. Examples of polycarboxylic acids that can be used to manufacture this type of polyester include dicarboxylic acids such as terephthalic acid,

orthophthalic acid, isophthalic acid, sodium sulfoisophthalate, 2,6-naphthalenedicarboxylic acid, 2,6-naphthalene dimethylene carboxylic acid, paraphenylene dicarboxylic acid, 1,4-cyclohexane dicarboxylic acid, cyclohexane diacetic acid, succinic acid, glutaric acid, adipic acid, suberic acid, azelaic acid, sebacic acid, and dodecanedioic acid; tricarboxylic acids such as trimellitic acid; and tetracarboxylic acids such as pyromellitic acid. One or more of these carboxylic acids can be used. However, the maximum amount of a carboxylic acid with three or more carboxyl groups is 0.5 mol%. It should be understood that these acids include the corresponding acid anhydrides, esters, and acid chlorides.

[0008] 2. Examples of suitable polyhydric alcohols include dihydric alcohols such as aliphatic/alicyclic diols represented by ethylene glycol, 1,2-propylene glycol, 1,4-butanediol, 1,5-pentanediol, neopentyl glycol, 1,6-hexanediol, diethylene glycol, triethylene glycol, polyethylene glycol (degree of polymerization up to 5), polytetramethylene glycol, 2,2-dimethyl-trimethylene glycol, hexamethylene glycol, p-xylene glycol, 1,4-cyclohexane dimethanol, neopentyl glycol, and ethylene oxide adducts of bisphenol A; trihydric and tetrahydric alcohols such as trimethylol propane and pentaerythritol; and the like. The maximum amount of alcohols with three or more hydroxyl radicals should be 0.5 mol% from the standpoint of moldability provided by the resulting polyester.

[0009] Many polycarboxylic acids and polyhydric alcohols are cited above, but the amorphous copolyester may also be obtained by means of using terephthalic acid together with a component that reduces polyester crystallinity (such as isophthalic acid, 2,6-naphthalenic acid, and 2,6-naphthalene dimethylene carboxylic acid) as the polycarboxylic acid. In this case, ethylene glycol or 1,4-cyclohexane dimethanol (CHDM) can be used alone as the glycol component. Moreover, amorphous copolyester can be obtained by means of using the crystallinity-reducing 1,4-cyclohexane dimethanol (CHDM) as the glycol component together with ethylene glycol. The latter copolyester is particularly preferred. In either case, it is important in terms of designing a copolyester for practical use that ester units from terephthalic acid be included as the polycarboxylic acid in the technique designed to improve product characteristics and based on the copolymerization of the above-mentioned polyester. Consequently, amorphous random copolymer resin can be obtained, for instance, by means of performing a cocondensation using a compound that imparts amorphous properties to the dicarboxylic acid and/or diol, with the ester units from the terephthalic acid and ethylene glycol being the principal component. Isophthalic

acid and 1,4-cyclohexane dimethanol can be cited as preferred compounds that impart such amorphous properties. A specific example of such a polymer is a copolyester that comprises ethylene terephthalate (ET) units and 1,4-cyclohexylene methylene terephthalate (CHDMT) units, particularly ET units > CHDMT units with an average molecular weight of approximately 26,000 (Mn) and an inherent viscosity IV of 0.75 (PET G6763; Eastman Chemical Company). [0010] B. The (meth)acrylic acid polymer that is admixed in order to improve foaming properties and calenderability in accordance with the present invention can be a methyl (meth)acrylate homopolymer or copolymer where methyl (meth)acrylate accounts for 50 to 95 wt% of the polymerizable components and where the copolymer component is, for instance, one of the following: a methacrylic acid ester of an alcohol with 2 to 18 carbons; an acrylic acid ester of an alcohol with 1 to 18 carbons; styrene,  $\alpha$ -methyl styrene, or another aromatic vinyl; acrylonitrile; methacrylonitrile; maleic acid; or itaconic acid. These copolymer components can be used alone or as a combination of two or more. Moreover, it is particularly preferred that the weightaverage molecular weight of these (meth)acrylic polymers be within a range of 500,000 to 5,000,000. Metablen® P501, 530, 531, 550, and 551 marketed by Mitsubishi Rayon Co., Ltd. can be cited as specific products.

#### [0011] C. Lubricant

The alkyl groups (or alkyl groups of the alkylphenyl groups) represented by R in the lubricant expressed by formula (I) have 6 to 20 carbons, and the alkyl groups of the alkyl polyoxyethylene and alkyl phenyl polyoxyethylene groups have 6 to 20 carbons, and particularly 9 or more carbons. A polyoxyethylene to which two or more oxyethylene units have been added can be cited as an example. The amount in which compound (I) is admixed is 0.01 to 5.0 parts by weight, and preferably 0.05 to 2.0 parts by weight, per 100 parts by weight of amorphous random copolyester resin. If this amount is less than 0.01 part by weight, the product will have an inadequate effect as a lubricant, and if more than 5.0 parts by weight are admixed, there will be a tendency toward plate-out during calendering, blooming and bleeding will be more likely to occur over time, and the resulting film will lose some of its transparency or the like. When the lubricant properties of mixtures combined with the lubricant represented by the above-mentioned formula (I) and used in the present invention were studied, it was confirmed that low-density oxidized and high-density oxidized polyethylene waxes (partially oxidized polyethylene with an acid value of 1 to 40 mg KOH/g and an average molecular weight of 10,000 or less) made the

lubricant of formula (I) more effective. Taking transparency of the resulting film into consideration, it is particularly preferred that the amount in which the above-mentioned polyethylene wax is admixed is 0.01 to 5.0 parts by weight per 100 parts by weight of amorphous random copolyester resin.

#### [0012] D. Plasticizer

The soft polyester composition that is the goal of the present invention cannot be obtained by means of admixing a small amount of plasticizer, and admixing at least 5 parts by weight of plasticizer is necessary. Consequently, the copolyester should be compatible with a large amount of plasticizer. However, there is a tendency toward bleeding when 40 parts by weight or more are added, and, therefore, the preferred range is 5 to 40 parts by weight. Moreover, a plasticizer that has good compatibility with the polymer is preferred. It is preferred that the SP (solubility parameter), which is to a certain extent an indicator of this compatibility, is 8.7 or higher, and particularly 9.0 or higher. In addition, in terms of chemical structure, a plasticizer with a structure similar to the components constituting the polyester is preferred. It is possible to use phthalic acid ester, aliphatic dibasic acid ester, trimellitic acid ester, epoxidized oil, or another plasticizer commonly used for polyvinyl chloride. Nevertheless, it was clarified from the results of many experiments that compounds having a polyalkylene oxide structure with ethylene oxide (-CH2-CH2-O-) units and/or popylene oxide (-CH2-CH(CH3)-O-) units, which were not previously used as plasticizers for polyester resins, are very effective as plasticizers that are compatible with copolyester. Ethylene glycol, diethylene glycol, triethylene glycol, polyethylene glycol, and the like, as well as ester or ether compounds thereof can be cited as preferred plasticizers with ethylene oxide (-CH<sub>2</sub>-CH<sub>2</sub>-O-) units. Similarly, propylene glycol, dipropylene glycol, and tripropylene glycol, as well as esters and ethers thereof can be cites as examples of plasticizers with propylene oxide (-CH<sub>2</sub>-CH(CH<sub>3</sub>)-O-) units. It is also apparent that polyalkylene glycol compounds comprising ethylene oxide (-CH-2-CH2-O-) units and propylene oxide (-CH<sub>2</sub>-CH(CH<sub>3</sub>)-O-) units, as well as polyethylene oxide and polypropylene oxide blocks, may also be useful as the plasticizer that constitutes the polyester composition of the present invention. Ethylene oxide adducts of poly(propylene oxide) glycol and copolymers of ethylene oxide and propylene oxide can be cited as substances having both of the above-mentioned structures. Alkylphenol ethylene oxide n-mole adducts and higher alcohol ethylene oxide nmole adducts can be cited as examples of such ether compounds, and aliphatic acid ethylene

oxide n-mole adducts and ethylene oxide n-mole adducts of aliphatic dibasic acid esters can be cited as examples of such ester compounds. Examples of other compounds having the above-mentioned units include ethylene oxide n-mole adducts of higher fatty acid amines, ethylene oxide n-mole adducts of bisphenol A, and other substances having the structure expressed by structural formula A. It should be noted that m in above-mentioned structural formula A is 1 to 20, and preferably 2 to 16.

[0013] Of the above-mentioned plasticizers, the preferred compound is one with a molecular weight of 300 to 900 and with an m-value of 2 or greater in structural formula A. Such a compound has excellent compatibility with the copolyester resin and is useful for enhancing plasticity. Moreover, in terms of chemical structure, ethers or esters of the above-mentioned polyalkylene oxide are preferred, ethers or esters of polyethylene oxide are further preferred, and monoethers or monoesters, such as polyoxyethylene monomethyl ether, polyethylene glycol monooleate, and polyethylene glycol C<sub>8</sub> acid esters, are ideal.

#### [0014] E. Foaming agent

Chemical forming agents, such as ADCA, OBSH, TSH (paratoluenesulfonyl hydrazide), and DPT (dinitropentamethylene tetramine), or mixed blends of these foaming agents are preferred as the foaming agents for the resin composition that forms the foaming soft polyester layer. These foaming agents can produce a foaming decorative sheet with good foamability as a result of being combined with the above-mentioned copolyester resin composition. In terms of the correlation with the softening temperature of the copolyester resin composition that is used, foamability is enhanced by means of reducing the temperature at which the foaming agent starts decomposing as a result of adding 0.5 to 1 part by weight of a foaming auxiliary agent (kicker), for instance, Zn. In particular, the softening temperature of the copolyester resin composition can be reduced by means of the addition of a plasticizer, and, therefore, a foaming auxiliary agent (kicker) may sometimes be necessary. However, the copolyester resin composition of the present invention has a broad softening temperature range suitable for foaming, and, therefore, a kicker usually is not necessary. The preferred foaming ratio is 2 to 20, and particularly 5 to 20. It is important that wallpaper be as lightweight as possible in order to provide good flame resistance as represented by the temperature-time area (corresponding to  $td\theta$ ) above the standard temperature curve defined in JIS A1321. Therefore, a foamed structure is useful for making wallpaper that is lightweight and still thick enough for appearance. Consequently, good

foamability is needed for materials that make wallpaper, and a foaming ratio of at least 2 is necessary. A foaming ratio of at least 5 is needed to realize a sense of volume. Nevertheless, currently cell roughness is extreme and sufficient commercial value cannot be realized if the foaming ratio is 20 or higher.

[0015] Calendering can be used for sheet (film) processing, and films that are 0.1 to 1.0 mm thick can be manufactured. The optimum temperature range for calendering is 100 to 220°C, and preferably 120 to 200°C. A foaming decorative sheet with a substrate layer can be manufactured by means of laminating the material under heat and pressure onto paper used for the wallpaper substrate. The film that is obtained from the soft copolyester resin composition to which a plasticizer has been added also has advantages in terms of properties associated with laminating by means of heating and pressing. Further decorative treatments, delustering treatments, and the like can be performed on the resulting foaming decorative sheet. For instance, printing (design printing), which is one means of decoration, can be accomplished by means of a method whereby the design is printed with printing ink made of an acrylic resin and/or urethane resin vehicles with the aid of the engraving roll of a gravure printing machine, and then the product is dried in a drying oven. Moreover, a surface treatment such as delustering can be carried out using a method whereby an agent for adjusting color is used by the same type of printing means as described above. The above-mentioned acrylic resin and/or urethane resin treatment agent can be used as a vehicle for the treatment agent. The foaming decorative sheet of the present invention is made of soft polyester and is preferred because of the good compatibility with the ink or treatment agent. Furthermore, the foaming decorative sheet of the present invention has good draw transfer properties.

#### [0016]

### [Working Examples] Working Example 1 and Comparative Examples

The plasticizer compounds, lubricants, calcium carbonate, foaming agents, and (meth)acrylic acid copolymers cited in Table 1 were admixed as cited in Table 1 with 100 parts by weight of the commercially available PET G6763 as the copolyester to prepare a polyester resin composition for calendering. The calendering conditions of the test roll are also shown in Table 1.

[0017] [Table 1]

		(1)	(2)
PET-G6763		100	100
_		parts	parts
RS-1000: Asahi Denka Co., Ltd.	RS-1000 Asahi Denka Co., Ltd.	22	22
Lubricant: AX-5181: Daikyo Kasei Co., Ltd.	Monostearyl phosphoric acid ester, distearyl phosphoric acid ester 6:4 mixture	0.8	0.8
Lubricant: Zinc stearate		1	1
R-101	Titanium oxide (rutile-type)	8	8
NS#100	Calcium carbonate	60	60
Foaming agent: #3170	ADCA	6.8	6.8
Methyl methacrylate copolymer	Metablen P530	0	1
Calendering conditions			
Roll temperature, °C		150	150
Number of revolutions (revolutions/minute)		30/25	30/25
Roll gap (m/m)		0.3	0.3
Foam cell status	Foamed 1000% to 1100%	Δ	0
Bank status (roughness of texture)		Δ	0
Elongation during when sheet exits		55 cm	55 cm
Condition of sheet selvedge		Δ	O-O <sub>∆</sub>

[0018] It is clear from Table 1 that calenderability is improved in (2), where Metablen P530 was added, in comparison with (1), in which the compound was not added. Furthermore, the condition of foam cells is improved and a product having good appearance is obtained, even with foaming by 1000% to 1100%.

#### [0019] Working Example 2

The relation with the flame resistance of the resulting sheet is shown in Fig. 1 for a case in which the amount in which calcium carbonate was admixed was varied. In Fig. 1, the  $td\theta$  on the ordinate axis has been measured according to JIS A1321, and it can be seen that flame resistance improves with a reduction in the measured values. The amount in which the composition is admixed is shown in Table 2. It is clear that flame resistance can be improved by means of increasing the amount in which calcium carbonate is admixed. Moreover, when the surface properties of the molded product were investigated, it was found that numerous irregularities had formed in the surface when 70 parts by weight of calcium carbonate were admixed.

## [0020]

#### [Table 2]

Pet-G 6763		100
RS-1000: Asahi Denka Co., Ltd.	Polyether ester	15
G-450: Nippon Oil & Fat Co., Ltd.	Polyoxyethylene glyceryl ether	5
Lubricant: AX-5181: Daikyo Kasei Kogyo Co., Ltd.	Monostearyl, distearyl phosphoric acid ester 6:4 mixture	0.5
R-101	Titanium oxide (rutile-type)	8
NS#100	Calcium carbonate	Amount of change: Fig. 1
Foaming Agent: #3170	ADCA	6.8
Metablen P530	Calendering auxiliary agent	1

#### [0021] Working Example 3

Fig. 2 shows the flame resistance of a sheet that was obtained by means of adding various amounts of aluminum hydroxide or antimony trioxide as a flame retardant (thickness of 0.15 mm). The amount in which the composition used for calendering was admixed is shown in Table 3.

#### [0022]

#### [Table 3]

Pet-G 6763		100
RS-1000: Asahi Denka Co., Ltd.	Polyether ester	15
G-450: Nippon Oil & Fat Co., Ltd.	Polyoxyethylene glyceryl ether	5
Lubricant: AX-5181: Daikyo Kasei Kogyo Co., Ltd.	Monostearyl, distearyl phosphoric acid ester 6:4 mixture	0.8
R-101	Titanium oxide (rutile-type)	8
NS#100	Calcium carbonate	Amount of change: Fig. 2
Flame retarder	Aluminum hydroxide	Amount of change: Fig. 2
Flame retarder	Antimony trioxide	Amount of change: Fig. 2
Foaming Agent: #3170	ADCA	6.8
Metablen P530	Calendering auxiliary agent	1

## [0023]

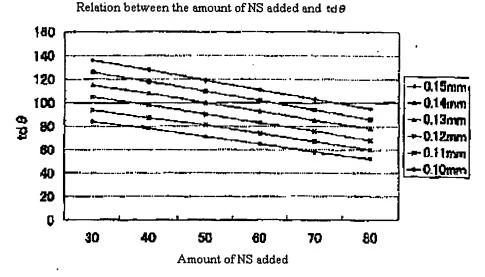
[Effects of the Invention] As was previously explained, wallpaper is produced by means of calendering after a (meth)acrylic polymer is admixed. The calenderability of the soft polyester is

considerably improved. In particular, the present invention has a considerable effect in that it is possible to add a large amount of calcium carbonate without compromising foamability, and, as a result, the flame resistance of the wallpaper is improved.

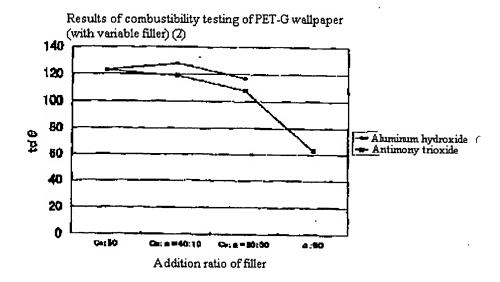
#### [Brief Description of the Drawings]

[Figure 1] Relation between the amount of calcium carbonate admixed (NS#100) and  $td\theta$  [Figure 2] Relation between  $td\theta$  and the joint use of calcium carbonate (Ca) and a flame retarder ( $\alpha$ ) (aluminum hydroxide, antimony trioxide)

[Fig. 1]



[Fig. 2]



# Continuation of front page

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